

Remarks

Claims 1-20 were pending.

Claims 9, 10 and 20 are canceled.

Claims 1, 6, 7, 16 and 17 are amended.

Claims 1-8 and 11-19 are now pending and are under consideration.

Claim 1 is amended to delete anhydrides as component d). Claim 1 is also amended to define epoxides of component d) according to original claim 11.

Claims 9 and 10 are canceled as not being further limiting.

Claim 6 is amended to require the presence of a phenolic antioxidant.

Claim 7 is amended to require the presence of a phosphorus processing stabilizer.

Claims 16 and 17 are amended to be more clear.

No new matter is added.

Claim 20 is rejected under 35 USC 112, second paragraph and 35 USC 101.

Claim 20 is canceled.

Claims 1-6, 8, 12, 13, 19 and 20 are rejected under 35 USC 102(b) as being anticipated by Kausch, et al., U.S. Pat. No. 6,414,070.

Kausch is cited as disclosing a polyolefin nanocomposite and maleic anhydride grafted polypropylene.

Anhydrides are now deleted as a group of component d). Thus, these rejections are addressed and are overcome.

Claims 1-9 and 12-20 are rejected under 35 USC 102(b) as being anticipated by Loontjens, et al., WO 01/05880.

Loontjens is cited as disclosing a polyolefin nanocomposite and glycidyl acrylate and anhydrides.

Present claim 1 is amended to include the limitations of claim 11 and to delete anhydrides as component d).

Applicants submit that in view of the claim amendments, that these rejections are addressed and are overcome.

Claims 8 and 9 are rejected under 35 USC 103(a) as being unpatentable over Kausch or Loontjens in view of JP 05-271481.

The Examiner cites that Kausch or Loontjens do not recite a polyfunctional epoxy compound. The Examiner must mean here that claims 10 and 11 are rejected.

Applicants respectfully rebut these rejections.

Applicants submit herewith an English translation of JP 05-271481. It is believed that no fee is required to submit this reference for consideration.

The instant invention is directed to the stabilization of thermoplastic polymers filled with nano-scaled clays. The clays are generally modified with long chain alkyl or dialkyl ammonium ions or amines or in a few cases onium ions such as phosphonium. The ammonium ion/amine additives are usually incorporated into the clay structure by a separate solution intercalation step. See the paragraph bridging pages 1 and 2 of the disclosure.

These organic modified clays have a number of disadvantages when used for the preparation of polyolefin nanocomposites. Ammonium salts are thermally unstable at temperatures used in polyolefin processing or may be otherwise reactive under processing conditions. These instabilities result in poor processing stability, inferior mechanical properties, discoloration, odor formation and reduced long term stability. See the first full paragraph, page 2 of the disclosure.

The inventors have surprisingly found that improved nanocomposites with an increased thermostability, with reduced odor and reduced undesired discoloration, which occurs as a result of the decomposition of the modification agents, can be prepared by the use of a mixture comprising a phenolic antioxidant and/or processing stabilizer and a mono or polyfunctional compound selected from the group of epoxides, oxazolines, oxazolones, oxazines, isocyanates and anhydrides, last full paragraph of page 2 of the disclosure.

Kausch and Loontjens disclose polyolefin nanocomposites.

JP '481 teaches the thermal improvement of conventionally filled polymers. The improvement is achieved by the addition of (pages 5 and 6 of the translation):

- C) a specific phenolic antioxidant;
- D) a sulphur containing antioxidant;
- E) a nitrogen containing antioxidant and
- F) a bisphenol A type epoxide.

To solve the problem of insufficient thermal stability for conventionally filled polymers, a combination of 4 components is necessary. The individual components are not sufficient, page 5, paragraphs 0003 and 0004. In particular, the addition of an epoxide alone leads to discoloration, page 5, paragraph 0004. In view of this disclosure, the skilled person would never contemplate epoxides as useful additives in combination with phenolic antioxidants in order to improve the thermostability of nano-scaled clay filled polyolefins.

It has been the merit of the inventors having surprisingly found that an improved long term stability in terms of color and heat resistance can be achieved for polymers which are filled with nano-scaled clay. The solution found is the combined use of a phenolic antioxidant and/or a processing

stabilizer and a compound selected from the class of the epoxides (as in amended claim 1), oxazolines, oxazolones, oxazines and isocyanates.

The results are surprising; the combinations show a significant improvement when compared to the individual stabilizers (Table 1, page 56). When the epoxide compound is added to the stabilizers, (Examples 1e, 1f and 1g), color is improved and heat stability is maintained, compared with the samples containing stabilizers alone. These results are surprising and could not have been expected based on the combined disclosures of the cited art.

In view of the present amendments and the above remarks, Applicants submit that the present 35 USC 103(a) rejections are addressed and are overcome.

In view of all of the above, Applicants submit that each of the claim rejections are addressed and are overcome.

The Examiner is kindly requested to reconsider and to withdraw the present rejections.

Applicants submit that the present claims are now in condition for allowance and respectfully request that they be found allowable.

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Attachment: English translation of JP 05-271481

PATENT LAID-OPEN (A)

No. H05-271481

October 19, 1993

Application No.:	H04-68133
Filing Date:	March 26, 1992
Applicant:	Sumitomo Chemical Co., Ltd.
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Int. Cl ⁵ :	C 08 L	23/00
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		5/13
		5/15
		5/17

(54) [Title of the Invention] POLYOLEFIN RESIN COMPOSITION

(57) [Abstract]

[Object] Specific compounds are incorporated into a synthetic polyolefin resin containing an inorganic filler to improve stability against thermal oxidation and hue.

[Constitution] A synthetic polyolefin resin (A) is mixed with (B) an inorganic filler, (C) a specific phenolic antioxidant such as 3,9-bis[2-{3-(3-t-butyl-4-hydroxy-5-methylphenyl)propionyloxy}-1,1-dimethylethyl]-2,4,8,10-tetraoxaspiro[5.5]undecane, (D) a specific sulfuric antioxidant such as distearyl 3,3'-thiodipropionate, (E) a specific nitrogen-containing compound such as ethylenebis(stearylamine), and (F) a bisphenol A type

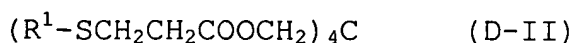
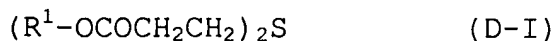
glycidyl ether epoxy compound.

[Effect] This composition exhibits excellent stability against thermal oxidation, as well as it imparts stable hue with less coloration.

[Scope of Claims for Patent]

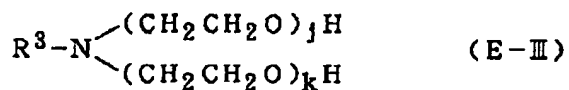
[Claim 1] A polyolefin resin composition comprising (A) a synthetic polyolefin resin, (B) an inorganic filler, (C) a phenolic antioxidant selected from 3,9-bis[2-{3-(3-t-butyl-4-hydroxy-5-methylphenyl)propionyloxy}-1,1-dimethylethyl]-2,4,8,10-tetraoxaspiro[5.5]undecane, 1,3,5-tris(3,5-di-t-butyl-4-hydroxybenzyl)isocyanurate and tetrakis[3-(3,5-di-t-butyl-4-hydroxyphenyl)propionyloxymethyl]methane, (D) a sulfuric antioxidant selected from the compound group represented by the following general formulas (D-I) and (D-II):

[Chemical formula 1]



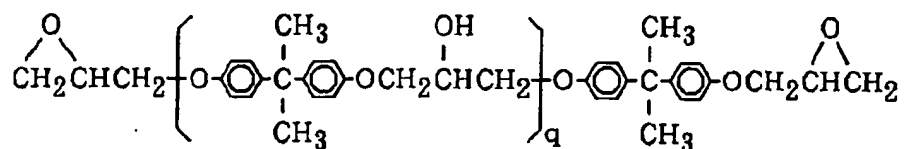
wherein R^1 represents alkyl having 4 to 20 carbon atoms, (E) a nitrogen-containing compound selected from the compound group represented by the following general formulas (E-I) to (E-III):

[Chemical formula 2]



wherein R^2 represents alkyl having 5 to 21 carbon atoms or alkenyl having 5 to 21 carbon atoms; n represents an integer from 1 to 6; R^3 represents alkyl having 8 to 18 carbon atoms, alkenyl having 8 to 18 carbon atoms, or acyl having 8 to 18 carbon atoms in total; j and k are each 1 or larger, while $j+k$ represents a number between 2 and 10, and (F) a bisphenol A type glycidyl ether epoxy compound represented by the following general formula (F-I):

[Chemical formula 3]



(F-I)

wherein q represents a number of 0 or larger.

[Claim 2] The resin composition according to claim 1, comprising 1 to 50% by weight of the inorganic filler (B) based on the weight of the synthetic polyolefin resin (A), and comprising 0.001 to 1 part by weight of the phenolic antioxidant (C), 0.002 to 2 parts by weight of the sulfuric antioxidant (D), 0.01 to 5 parts by weight of the nitrogen-containing compound (E), and 0.01 to 5 parts by weight of the epoxy compound (F), based on 100 parts by weight of the sum of the synthetic polyolefin resin (A) and inorganic filler (B).

[Detailed Description of the Invention]

[0001]

[Field of Application in Industry] The present invention relates to a polyolefin resin composition having an inorganic filler incorporated therein, and more particularly, to a polyolefin resin composition exhibiting excellent stability against thermal oxidation and at the same time, excellent hue.

[0002]

[Prior Art] Polyolefin resins including polyethylene and polypropylene have excellent physical, chemical and electrical properties, and are therefore used widely. However, for the purpose of improving the mechanical properties such as toughness, impact resistance, dimensional stability and thermal resistance, a technique of incorporating an inorganic filler such as talc is being employed. On the other hand, since polyolefin resins are

susceptible to deterioration due to thermal oxidation, oxidation preventive measures such as the use of a phenolic antioxidant and a sulfuric antioxidant in combination, have been traditionally taken. However, polyolefin resins containing incorporated inorganic fillers have a drawback that the stability against thermal oxidation is significantly deteriorated because the antioxidant adsorbs to the inorganic filler, and thus an improvement is highly demanded.

[0003] Thus, in order to improve stability against the thermal oxidation of polyolefin resins having incorporated inorganic fillers, a method of incorporating an epoxy compound (Japanese Unexamined Patent Publication No. Hei 1-188543), a method of incorporating an amide or amine compound (Japanese Unexamined Patent Publication No. Hei 2-49042) and the like have been suggested.

[0004]

[Problems to be Solved by the Invention] However, in these known methods, sufficient stability against thermal oxidation could not be necessarily obtained. Further, in the case of using an epoxy compound, there were problems such as discoloration of the resin composition. The inventors of the present invention conducted various studies in order to solve such problems and develop a polyolefin resin composition having excellent stability against thermal oxidation and at the same time having excellent hue, thus completing the present invention.

[0005]

[Means for Solving the Problems] Thus, the present invention is to provide a polyolefin resin composition containing (A) a synthetic polyolefin resin, (B) an inorganic filler, and the following components (C) to (F):

[0006] (C) A phenolic antioxidant selected from the

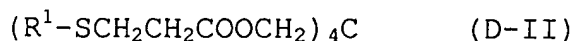
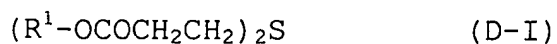
following group of compounds:

3,9-bis[2-{3-(3-t-butyl-4-hydroxy-5-methylphenyl)propionyloxy}-1,1-dimethylethyl]-2,4,8,10-tetraoxaspiro[5.5]undecane, 1,3,5-tris(3,5-di-t-butyl-4-hydroxybenzyl)isocyanurate, and tetrakis[3-(3,5-di-t-butyl-4-hydroxyphenyl)propionyloxymethyl]methane,

[0007] (D) A sulfuric antioxidant selected from the group of compounds represented by the following general formulas (D-I) and (D-II):

[0008]

[Chemical formula 4]

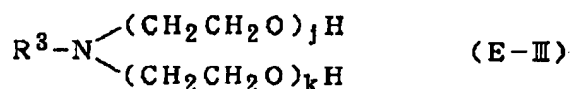
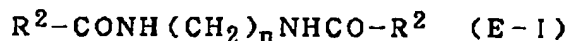


[0009] wherein R^1 represents alkyl having 4 to 20 carbon atoms,

[0010] (E) A nitrogen-containing compound selected from the group of compounds represented by the following general formulas (E-I) to (E-III):

[0011]

[Chemical formula 5]



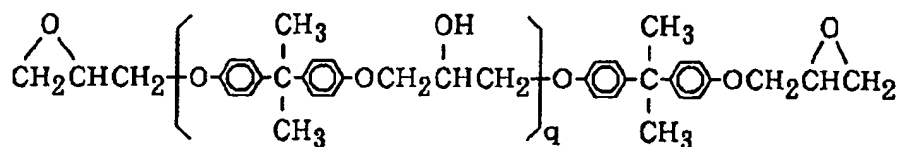
[0012] wherein R^2 represents alkyl having 5 to 21 carbon atoms, or alkenyl having 5 to 21 carbon atoms; n represents an integer from 1 to 6; R^3 represents alkyl having 8 to 18 carbon atoms, alkenyl having 8 to 18 carbon atoms, or acyl having 8 to 18 carbon atoms in total; and j and k are each 1 or larger, while $j+k$ represents a number between 2 and 10, and

[0013] (F) A bisphenol A type glycidyl ether epoxy

compound represented by the following general formula (F-I):

[0014]

[Chemical formula 6]



(F-I)

[0015] wherein q represents a number of 0 or larger.

[0016] The synthetic polyolefin resins (A) used in the present invention include homopolymers of α -olefin such as ethylene, propylene, 1-butene, 1-hexene and 4-methyl-1-pentene, random and block copolymers formed from two or more kinds of α -olefin, copolymers of an α -olefin and another copolymerizable unsaturated compound, and the like. These may be used individually, or as a blend of two or more kinds.

[0017] Examples of the homopolymers of α -olefin include polyethylene, polypropylene, poly-1-butene, polyisobutene, poly-3-methyl-1-butene, poly-4-methyl-1-pentene, and the like. Examples of the copolymers of α -olefins include ethylene/propylene copolymers, ethylene/1-butene copolymers, propylene/4-methyl-1-pentene copolymers, propylene/1-butene copolymers, 1-decene/4-methyl-1-pentene copolymers, ethylene/propylene/1-butene copolymers, and the like. Furthermore, those containing a polyunsaturated compound such as conjugated diene or non-conjugated diene, or a monounsaturated compound such as acrylic acid, methacrylic acid or vinyl acetate, together with an α -olefin, as the copolymer components, are also included. These polymers may be acid-modified polymers, for example,

polymers graft-modified with an α,β -unsaturated fatty acid, an alicyclic carboxylic acid or a derivative thereof.

[0018] In the present invention, blends formed by mixing these synthetic polyolefin resins with synthetic rubber, can also be used in accordance with the use. Examples of the synthetic rubber which is suitable for the blends include ethylene/ α -olefin copolymer rubbers, and the like. The ethylene/ α -olefin copolymer rubbers include copolymer rubbers of ethylene and another α -olefin, for example, propylene, 1-butene, 1-hexene or the like; ternary copolymer rubbers formed by copolymerizing an ethylene/propylene system with a non-conjugated diene as the third component, for example, ethylidenenorbornene, dicyclopentadiene or the like.

[0019] In the composition having an inorganic filler incorporated therein, as in the present invention, particularly propylene-based synthetic resins, for example, polypropylene, copolymers of propylene with another α -olefin, or blends of these and a synthetic rubber are preferably used as the base resin.

[0020] The inorganic fillers (B) used in the present invention include talc, mica, carbon black, titanium oxide, zinc oxide, aluminum hydroxide, calcium carbonate, magnesium carbonate, calcium sulfate, barium sulfate, calcium silicate, magnesium silicate, celite, kaolin, zeolite, silica, asbestos, glass fiber, carbon fiber, barium titanate, lead titanate, and the like. These can be used individually, or in combination or two or more kinds. Among these, for example talc, mica or glass fiber is preferably used from the viewpoint of improving low temperature impact resistance, moldability and coating properties.

[0021] According to the present invention, a specific phenolic antioxidant (C), a specific sulfuric antioxidant (D), a specific nitrogen-containing compound (E) and a specific epoxy compound (F) are further mixed with the synthetic polyolefin resin (A) and inorganic filler (B).

[0022] The phenolic antioxidant of the component (C) is selected from the following group of compounds, and these may be used individually, or may be used in combination of two or more kinds.

[0023] 3,9-bis[2-{3-(3-t-butyl-4-hydroxy-5-methylphenyl)propionyloxy}-1,1-dimethylethyl]-2,4,8,10-tetraoxaspiro[5.5]undecane, 1,3,5-tris(3,5-di-t-butyl-4-hydroxybenzyl)isocyanurate, and tetrakis[3-(3,5-di-t-butyl-4-hydroxyphenyl)propionyloxymethyl]methane.

[0024] The sulfuric antioxidant of the component (D) is represented by the general formula (D-I) or (D-II) described above. In the general formulas (D-I) and (D-II), R^1 is alkyl having 4 to 20 carbon atoms, and a preferred number of carbons for the alkyl is 6 to 18. These sulfuric antioxidants may also be used individually, or may be used in combination of two or more kinds. Specific examples of the sulfuric antioxidant (D) used in the present invention include the following compounds.

[0025] Dilauryl 3,3'-thiodipropionate, dimyristyl 3,3'-thiodipropionate, distearyl 3,3'-thiodipropionate, tetrakis(3-laurylthiopropionyloxymethyl)methane.

[0026] The nitrogen-containing compound of the component (E) is represented by any one of the general formulas (E-I) to (E-III) described above.

[0027] In the general formulas (E-I) and (E-II), R^2 is alkyl or alkenyl, and the number of carbon atoms for each is 5 to 21. Among these, one having more carbon atoms, for example, alkyl having 11 to 21 carbon atoms or alkenyl

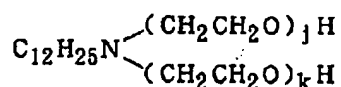
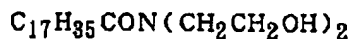
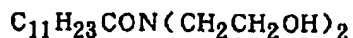
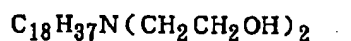
having 11 to 21 carbon atoms is preferred. Furthermore, in the general formula (E-I), n is an integer from 1 to 6.

[0028] In the general formula (E-III), R^3 is alkyl, alkenyl or acyl, and here, the number of carbon atoms for alkyl or alkenyl, and the number of carbon atoms in total for acyl are respectively 8 to 18. The acyl represented by R^3 is preferably alkanoyl or alkenoyl represented by R^4CO- , wherein R^4 is alkyl having 7 to 17 carbon atoms or alkenyl having 7 to 17 carbon atoms. R^3 is particularly preferably alkyl or alkanoyl. Furthermore, in the general formula (E-III), j and k are each 1 or larger, and are selected so that $j+k$ falls within the range of 2 to 10. The compounds of the general formula (E-III) in which j and k are each 1, are usually obtained as single products, but the compounds in which $j+k$ is greater than 2, are generally obtained as compounds having a constant value for $j+k$.

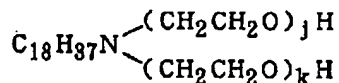
[0029] Specific examples of the compound represented by the general formula (E-I) include methylenebis(stearylamide), ethylenebis(stearylamide), ethylenebis(oleylamide), hexamethylenebis(stearylamide), and the like. Specific examples of the compound represented by the general formula (E-II) include lauric acid amide, stearic acid amide, oleic acid amide, behenic acid amide, erucic acid amide, and the like. Specific examples of the compound represented by the general formula (E-III) include the following.

[0030]

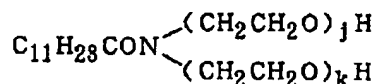
[Chemical formula 7]



$$(j+k=4, 8 \text{ or } 10)$$



$$(j+k=4, 8 \text{ or } 10)$$



$$(j+k=4, 8 \text{ or } 10)$$

[0031] These nitrogen-containing compounds (E) represented by general formula (E-I), (E-II) or (E-III) are all effective for the present invention, and can be used individually or in combination of two or more kinds. Particularly, in the present invention, ethylenebis(stearylamide), erucic acid amide or N,N-bis(2-hydroxyethyl)stearylamine is preferably used.

[0032] The epoxy compound of the component (F) represented by the general formula (F-I) is generally produced by condensation reaction between bisphenol A and epichlorohydrin. Therefore, the epoxy compound (F) is typically a group of molecules having a number (q) of repeating units. Furthermore, there are epoxy compounds (F) having higher molecular weights, obtained by first purifying a condensation reaction product of bisphenol A and epichlorohydrin to make the range of q narrower, and then subjecting the purified product to further condensation reaction with bisphenol A and epichlorohydrin.

[0033] There are commercially available compounds having q in the general formula (F-I) equal to or greater than 0, and up to about 35 on the average, and the products are in the solid state or in the liquid state at normal

temperature, but they all can be used for the present invention. Of course, these may be used individually, or in combination of two or more kinds. Such epoxy compounds generally show differences depending on the epoxy equivalent, and in the present invention, those having an epoxy equivalent of about 170 to about 5000 are preferably used. If available, a compound having an epoxy equivalent exceeding about 5000, or a compound in which the value of q in the general formula (F-I) exceeds about 35 on the average, can also be used for the present invention.

[0034] The mixing ratios for the respective components are not particularly limited in the present invention, but generally, for the synthetic polyolefin resin (A) and inorganic filler (B), it is preferable that the mixing ratio of the inorganic filler (B) relative to the synthetic polyolefin resin (A) is 1 to 50% by weight, and more preferably, the inorganic filler (B) is used at a ratio of 5 to 40% by weight. If the amount of the inorganic filler (B) is less than 1% by weight based on the synthetic polyolefin resin (A), the amount is insufficient for improving the mechanical properties such as toughness, dimensional stability and thermal resistance. If the amount exceeds 50% by weight, problems may arise in the moldability or the like.

[0035] The preferable amounts to be used for the respective components of (C) to (F) may be, based on 100 parts by weight of the sum of the synthetic polyolefin resin (A) and inorganic filler (B), 0.001 to 1 part by weight of the phenolic antioxidant (C), 0.002 to 2 parts by weight of the sulfuric antioxidant (D), 0.01 to 5 parts by weight each of the nitrogen-containing compound (E) and the epoxy compound (F). If the phenolic antioxidant (C) is used in an amount of less than 0.001 parts by weight, the

sulfuric antioxidant less than 0.002 parts by weight, the nitrogen-containing compound (E) less than 0.01 parts by weight, or the epoxy compound less than 0.01 parts by weight based on 100 parts by weight of the sum of the synthetic polyolefin resin (A) and inorganic filler (B), it is difficult to attain the desired effects sufficiently. Further, if the components are mixed in the amounts exceeding 1 part by weight, 2 parts by weight, 5 parts by weight and 5 parts by weight, respectively, effects corresponding to the amounts are not obtained, and it is economically inefficient.

[0036] The polyolefin resin composition of the present invention may include other additives, for example, processing stabilizers, antioxidants, photostabilizers, metal deactivators, metal soaps, nucleating agents, antistatic agents, lubricants, flame retardants, releasing agents, antifungal agents, pigments and the like, to be used in combination, as long as the additives do not impair the properties of the composition.

[0037] Specific examples of the processing stabilizer that can be used include the following.

[0038] 2-t-butyl-6-(3-t-butyl-2-hydroxy-5-methylbenzyl)-4-methylphenyl acrylate, 2-[1-(2-hydroxy-3,5-di-t-pentylphenyl)ethyl]-4,6-di-t-pentylphenyl acrylate.

[0039] The antioxidant may be exemplified by phenolic antioxidants other than the component (C), sulfuric antioxidants other than the component (D), and phosphoric antioxidants. Specific examples of the phosphoric antioxidants include the following.

[0040] Tris(nonylphenyl) phosphite, tris(2,4-di-t-butylphenyl) phosphite, bis(2,4-di-t-butylphenyl)pentaerythritol diphosphite, bis(2,6-di-t-butyl-4-methylphenyl)pentaerythritol diphosphite,

distearyl-pentaerythritol diphosphate, tetrakis(2,4-di-*t*-butylphenyl) 4,4'-biphenylenediphosphonite, 2,2'-ethylidenebis(4,6-di-*t*-butylphenyl) fluorophosphite, 2,2'-methylenebis(4,6-di-*t*-butylphenyl) octyl phosphite.

[0041] Examples of the photostabilizers include ultraviolet absorbents such as benzotriazoles, benzophenones, hydroxybenzoates and cyanoacrylates, nickel-based quenchers and hindered amine-based photostabilizers.

[0042] Specific examples of the ultraviolet absorbents include the following.

[0043] 2-(3-*t*-butyl-2-hydroxy-5-methylphenyl)-5-chlorobenzotriazole, 2-(3,5-di-*t*-butyl-2-hydroxyphenyl)benzotriazole, 2-(2-hydroxy-5-methylphenyl)benzotriazole, 2-(2-hydroxy-5-*t*-octylphenyl)benzotriazole, 2-(3,5-di-*t*-amyl-2-hydroxyphenyl)benzotriazole, 2-[2-hydroxy-3-(3,4,5,6-tetrahydrophthalimidemethyl)-5-methylphenyl]benzotriazole, 2-[2-hydroxy-3,5-bis(α , α -dimethylbenzyl)phenyl]benzotriazole, 2-hydroxy-4-octyloxybenzophenone, 2-hydroxy-4-methoxybenzophenone, 2,4-di-*t*-butylphenyl 3,5-di-*t*-butyl-4-hydroxybenzoate, ethyl 2-cyano-3,3-diphenylacrylate.

[0044] Specific examples of the hindered amine photostabilizers include the following.

[0045] Bis(2,2,6,6-tetramethyl-4-piperidyl) sebacate, bis(1,2,2,6,6-pentamethyl-4-piperidyl) sebacate, 2-methyl-2-(2,2,6,6-tetramethyl-4-piperidyl) amino-N-(2,2,6,6-tetramethyl-4-piperidyl) propionamide, bis(1,2,2,6,6-pentamethyl-4-piperidyl) 2-(3,5-di-*t*-butyl-4-hydroxybenzyl)-2-*n*-butylmalonate, tetrakis(2,2,6,6-tetramethyl-4-piperidyl) 1,2,3,4-butanetetracarboxylate, poly[{6-(1,1,3,3-tetramethylbutyl)imino-1,3,5-triazine-2,4-diyl} {(2,2,6,6-tetramethyl-4-piperidyl)imino}hexamethylene

{(2,2,6,6-tetramethyl-4-piperidyl)imino}}, poly[(6-morpholino-1,3,5-triazine-2,4-diyl){(2,2,6,6-tetramethyl-4-piperidyl)imino}hexamethylene {(2,2,6,6-tetramethyl-4-piperidyl)imino}}, a polycondensate of dimethyl succinate and 1-(2-hydroxyethyl)-4-hydroxy-2,2,6,6-tetramethylpiperidine, a condensate of N,N'-bis(3-aminopropyl)ethylenediamine and 2,4-bis[N-butyl-N-(1,2,2,6,6-pentamethyl-4-piperidyl)amino]-6-chloro-1,3,5-triazine, 1,2,2,6,6-pentamethyl-4-piperidinol, and a condensate of 3,9-bis(2-hydroxy-1,1-dimethylethyl)-2,4,8,10-tetraoxaspiro[5.5]undecane and 1,2,3,4-butanetetracarboxylic acid.

[0046] Specific examples of the metal deactivators include the following.

[0047] N,N'-diphenyloxalic acid diamide, N-salicylidene N'-salicyloylhydrazine, N,N'-bis(salicyloyl)hydrazine, N,N'-bis[3-(3,5-di-t-butyl-4-hydroxyphenyl)propionyl]hydrazine, 3-salicyloylamino-1,2,4-triazole, bis(benzylidene)oxalic acid dihydrazide.

[0048] Specific examples of the nucleating agents include the following.

[0049] Alkali metal salts or alkaline earth metal salts such as sodium salts or calcium salts of aromatic monocarboxylic acids such as benzoic acid, toluic acid and p-t-butylbenzoic acid; dibenzylidenesorbitols such as 1,3·2,4-di(benzylidene)sorbitol, 1,3·2,4-di(p-methylbenzylidene)sorbitol and 1,3·2,4-di(p-ethylbenzylidene)sorbitol; metal salts of aromatic phosphoric acid compounds, such as sodium bis(4-t-butylphenyl)phosphate and sodium 6,6'-methylenebis(2,4-d-t-butylphenyl)phosphate.

[0050] Specific examples of the flame retardants include the following.

[0051] Phosphoric acid esters such as tricresyl phosphate, triphenyl phosphate, diphenyloctyl phosphate and tributyl phosphate; halogen-containing phosphoric acid esters such as tris(2-chloroethyl) phosphate and tris(2,3-dibromopropyl) phosphate; halogen-containing vinyl compounds such as vinyl chloroacetate, bromostyrene and bromophenyl allyl ether; halogen compounds such as chlorinated paraffin, brominated polyphenyl, perchloropentacyclodecane, tetrabromoethane, hexabromocyclododecane, tetrachlorophthalic anhydride, tetrabromophthalic anhydride, tetrabromobisphenol A and decabromodiphenyl oxide; metal-containing inorganic compounds such as antimony trioxide and aluminum hydroxide.

[0052] Specific examples of the pigment include the following.

[0053] Inorganic pigments such as carbon black, titanium white, titanium yellow, iron oxide and Cadmium Red; polycyclic organic pigments such as phthalocyanines, quinacridones, perylenes, anthraquinones and isoindolinones.

[0054]

[EXAMPLES] Hereinafter, the present invention will be described in more detail by way of Examples, but the present invention is not limited to these. In addition, the parts and % described in the Examples indicate parts by weight and % by weight, respectively, unless otherwise indicated.

[0055] The test specimen used in the Examples are as described in the following Tables, and hereinafter, the compounds will be indicated by the respective symbols.

[0056]

[Table 1] Inorganic filler

B-1: Talc [Micronwhite 5000S (trade name); manufactured by

Hayashi-Kasei Co., Ltd.]

B-2: Mica [S-150 (trade name); manufactured by Repco Co., Ltd.]

[0057]

[Table 2] Phenolic antioxidant

C-1: 3,9-bis[2-{3-(3-t-butyl-4-hydroxy-5-methylphenyl)propionyloxy}-1,1-dimethylethyl]-2,4,8,10-tetraoxaspiro[5.5]undecane

C-2: 1,3,5-tris(3,5-di-t-butyl-4-hydroxybenzyl)isocyanurate

C-3: Tetrakis[3-(3,5-di-t-butyl-4-hydroxyphenyl)propionyloxymethyl]methane

C-4: 2,2'-thiodiethylenebis[3-(3,5-di-t-butyl-4-hydroxyphenyl) propionate]

C-5: Triethylene glycol bis[3-(3-t-butyl-4-hydroxy-5-methylphenyl) propionate]

[0058]

[Table 3] Sulfuric antioxidant

D-1: Distearyl 3,3'-thiodipropionate

D-2: Tetrakis(3-laurylthiopropionyloxymethyl)methane

[0059]

[Table 4] Nitrogen-containing compound

E-1: Ethylenebis(stearylamide)

E-2: Erucic acid amide

E-3: N,N-bis(2-hydroxyethyl)stearylamine

[0060]

[Table 5] Epoxy compound

F-1: Bisphenol A type glycidyl ether epoxy compound
[Epikote 1002 (trade name); manufactured by Yuka Shell Epoxy Co., Ltd., epoxy equivalent: 600 to 700]

F-2: Bisphenol A type glycidyl ether epoxy compound
[Epikote 1007 (trade name); manufactured by Yuka Shell Epoxy Co., Ltd., epoxy equivalent: 1750 to 2200]

F-3: o-Cresol novolac type epoxy compound [Epikote 180S65 (trade name): Yuka Shell Epoxy Co., Ltd., epoxy equivalent: 205 to 220].

[0061] Compositions were prepared at the composition ratios indicated in Table 6 to Table 9, using an unstabilized propylene-ethylene block copolymer (ethylene content: 7.3%). In the tables, the amounts of the propylene/ethylene block copolymer and the inorganic filler are expressed in their respective ratios (%) based on the total weight of both components, and the amounts of the other components are expressed in their respective ratios (parts) based on 100 parts of the sum of the block copolymer and the inorganic filler. Each of the compositions was mixed with a Henschel mixer, and then kneaded with a 30-mm ϕ twin-screw extruder (manufactured by Nakatani Kikai Co., Ltd., NAS30 type, cylinder temperature: 230 to 240°C), to palletize the composition. Using these pellets, sheets having a size of 40×60×1 mm were molded by a 1-ounce injection molding machine (manufactured by Sanjo Seiki Co., Ltd., SAV-30A Model, cylinder temperature: 220 to 240°C).

[0062] A test piece having a size of 40×40×1 mm was produced from the obtained sheet, and evaluation was performed on the stability to thermal oxidation and the hue of the test piece. The stability to thermal oxidation was determined by placing the specimen in a gear oven at 150°C, and measuring the time taken until embrittlement occurs in the test piece (embrittlement life). A longer embrittlement life means more excellent stability to thermal oxidation. Furthermore, the hue was determined through the yellowness index (YI) at the surface of the test piece after molding. A smaller YI means more

excellent hue. The results are shown in Table 6 to Table 9.

[0063]

[Table 6]

Examples of the present invention

Run No.	Composition ratio						Test results	
	Block copolymer	In-organic filler	Phenolic anti-oxidant	Sul-furic anti-oxidant	Nitrogen-containing compound	Epoxy compound	150°C embrittlement life	Hue
	%	Type %	Upper line: type Lower line: parts				(hr)	(YI)
1	80	B-1 20	C-1 0.02	D-1 0.04	E-1 0.15	F-1 0.15	555	4.8
2	80	B-1 20	C-2 0.02	D-1 0.04	E-1 0.15	F-1 0.15	545	6.5
3	80	B-1 20	C-3 0.02	D-1 0.04	E-1 0.15	F-1 0.15	530	5.5
4	80	B-1 20	C-1 0.02	D-2 0.04	E-1 0.15	F-1 0.15	795	5.0
5	80	B-1 20	C-1 0.02	D-1 0.04	E-2 0.15	F-1 0.15	540	7.2

[0064]

* * [Table 7]

Examples of the present invention (continued)

6	80	B-1 20	C-1 0.02	D-1 0.04	E-3 0.15	F-1 0.15	460	4.9
7	80	B-1 20	C-1 0.02	D-1 0.04	E-1 0.15	F-2 0.15	550	4.3
8	60	B-1 40	C-1 0.04	D-1 0.08	E-1 0.25	F-1 0.25	815	5.7
9	80	B-2 20	C-1 0.02	D-1 0.04	E-1 0.15	F-1 0.15	450	5.6

[0065]

[Table 8]

Comparative Examples

Run No.	Composition ratio						Test results	
	Block copolymer	In-organic filler	Phenolic anti-oxidant	Sulfuric anti-oxidant	Nitrogen-containing compound	Epoxy compound	150°C embrittlement life	Hue
	%	Type %	Upper line: type Lower line: parts				(hr)	(YI)
11	80	B-1 20	C-4 0.02	D-1 0.04	E-1 0.15	F-1 0.15	270	5.8
12	80	B-1 20	C-5 0.02	D-1 0.04	E-1 0.15	F-1 0.15	290	5.0
13	80	B-1 20	C-1 0.02	D-1 0.04	E-1 0.3	-	390	3.8
14	80	B-1 20	C-2 0.02	D-1 0.04	E-1 0.3	-	150	4.3
15	80	B-1 20	C-3 0.02	D-1 0.04	E-1 0.3	-	350	4.8

[0066]

* * [Table 9]

Comparative examples (continued)

16	80	B-1 20	C-1 0.02	D-1 0.04	-	F-1 0.3	385	19.4
17	80	B-1 20	C-2 0.02	D-1 0.04	-	F-1 0.3	395	26.3
18	80	B-1 20	C-3 0.02	D-1 0.04	-	F-1 0.3	370	22.5
19	80	B-1 20	C-1 0.02	D-1 0.04	E-2 0.3	-	380	6.6
20	80	B-1 20	C-1 0.02	D-1 0.04	E-3 0.3	-	110	3.8
21	80	B-1 20	C-1 0.02	D-1 0.04	-	F-2 0.3	145	20.4
22	80	B-1 20	C-1 0.02	D-1 0.04	E-1 0.15	F-3 0.15	435	15.6

[0067]

[Effects of the Invention] According to the present invention, a composition formed by mixing a specific compound to a synthetic polyolefin resin containing an

inorganic filler exhibits excellent stability to thermal oxidation, and at the same time, gives stable hue with less coloration.